

The Effect of Electrical Boundary Conditions on the Thermal Properties of Ferroelectric Piezoelectric Ceramics

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The thermal conductivity of polycrystalline bulk PZT (lead-zirconate-titanate) has been investigated according to electrical boundary conditions and poling. The thermal conductivity of poled PZT was measured in the poling direction for open circuit and short circuit conditions. The short circuit thermal conductivity had the largest thermal conductivity. The relationship between these two thermal properties, the “electrothermal” coupling factor k_{33}^{κ} , was found to be similar to the electromechanical coupling factor k_{33} relating elastic compliance under short circuit and open circuit conditions. The thermal conductivity of the unpoled sample was found to have the lowest thermal conductivity. The significance of the thermal conductivity with regards to phonon mode scattering and elastic compliance was discussed.

Piezoelectric materials are a unique class of materials in which the electrical and mechanical properties are coupled. The most popular piezoelectric material is lead-zirconate-titanate (PZT) due to its large electromechanical properties and the adaptability of its properties with dopants.

It is well known that the electrical boundary conditions and poling affect the elastic compliance of piezoelectric materials.¹ The thermal properties, namely thermal conductivity, may also be expected to be affected by electrical boundary conditions and poling due to the fact that this property and elastic compliance both arise primarily from phonon mode phenomena.

The thermal properties of PZT ceramics have been studied at low temperature (20K-300K), and a transition temperature was found between 50K and 80K.² It has also been studied at high temperature, between 300K-800K, which characterizes the affect of phase transition on the thermal properties.³ However, the relationship of thermal properties with electrical boundary conditions and poling in PZT and other ferroelectric piezoelectric ceramics has not been studied.

Using the experiment outlined in⁴, which uses a time constant formulation to determine thermal properties, the thermal diffusivity α of a poled and depoled commercially available hard PZT ceramic discs, APC 841 (APC Int., USA), of a diameter of 51 mm was measured. The thermal diffusivity was measured in the direction of polarization for the poled samples. Using the DSC Q2000 (TA Instruments), the absolute value heat capacity of a small sample was measured by comparing it to a sapphire reference sample. The heat capacity and density are respectively $c_p = 340\text{J/kg K}$ $\rho = 7600\text{kg/m}^3$. The heat capacity is not affected by electrical boundary conditions and it is a scalar property; therefore, the thermal conductivity κ can be determined from the thermal diffusivity α and from the heat capacity c_p

$$\kappa = \alpha c_p \rho. \quad (1)$$

The original report regarding the TC thermal diffusivity experiment presented a standard deviation of less than 5%. However, the results of this experiment had larger deviation (Tab. 1) due to the fact that the diameters of the samples were a few millimeters smaller than the sample holder. The error found is different for different boundary conditions and poling states, but this is believed to be random.

The measured thermal diffusivity and thermal conductivity values for poled open circuit, poled short circuit, and depoled samples are described in Tab. I as an average of two measurements on three samples each. samples were depoled by heating them and then checking their response on a d_{33} meter. The short circuit thermal conductivity κ_{33}^E is more than 1.5 times larger than the open circuit one κ_{33}^D . The unpoled thermal conductivity κ^u showed the smallest value, 15% less than that of the open circuit case. The relationship between the open circuit κ_{33}^D and short circuit thermal conductivity κ_{33}^E can be described by a electrothermal coupling coefficient

$$\kappa_{33}^E (1 - (k_{33}^{\kappa})^2) = \kappa_{33}^D, \quad (2)$$

which is closely related to the relationship¹ between short circuit and open circuit elastic compliance and the electromechanical coupling coefficient k_{33}

$$s_{33}^E (1 - k_{33}^2) = s_{33}^D. \quad (3)$$

The electromechanical coupling factor of this ceramic found from electrical impedance spectroscopy is $k_{33} = 0.68$. Using Eq. 2, the “electrothermal” coupling factor can be calculated to be $k_{33}^{\kappa} = 0.63$. The error between the two coupling factors may be due to

Table I. Thermal diffusivity and thermal conductivity depending on electrical boundary conditions

| | Thermal diff. \pm (mm ² /s) | | Thermal cond. \pm (W/m K) | |
|---------------|--|------|-----------------------------|------|
| Open circuit | 0.50 | 0.02 | 1.4 | 0.06 |
| Short circuit | 0.82 | .08 | 2.3 | 0.23 |
| Depoled | 0.43 | 0.03 | 1.2 | 0.01 |

error in the thermal measurements and possibly other microscopic features which do not correlate between the k value determined from electrical and thermal measurements.

In summary, $\kappa_{33}^E > \kappa_{33}^D > \kappa^u$. This may be understood from phonon mode scattering, orientation of domains, and elastic compliance. Because of the random orientation of domains in the depoled sample, it is expected that there will be the most phonon scattering in this material. Therefore, it will have the lowest thermal conductivity. Because the domains of the poled material are oriented, less scattering is expected and thermal conductivity will be larger for the poled material (κ_{33}^E and κ_{33}^D). The elastic compliance under short circuit conditions s_{33}^E is softer than the elastic compliance under open circuit conditions s_{33}^D . This means that the lattice and domain wall motion are larger in the short circuit condition. The larger lattice vibration and domain motion in short circuit conditions will also correlate to a larger thermal conductivity in short circuit conditions κ_{33}^E due to increased phonon mode transport. This observation can also be used to understand the relation between k_{33} and k_{33}^κ .

The clear result of the experiments is that thermal conductivity in ferroelectric ceramics depends on electrical boundary conditions. It is very possible that the electromechanical coupling factor in these materials is related to thermal properties as well, namely thermal conductivity and thermal diffusivity. A discussion was presented to explain the behavior using phonon

scattering and domain orientation concepts. Future work includes studying the effect of microstructure on phonon mode transport in these materials and further clarifying the thermal-electrical coupling experimentally demonstrated in the experiments.

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